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Using self-phase modulation for temporal compression of intense femtosecond laser pulses

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Abstract. **We report a theoretical analysis of the spectrum broadening of intense chirped femtosecond pulses in media with cubic nonlinearity. Subsequent temporal compression of such pulses using a quadratic spectral phase corrector is investigated. Pulse compression from 57 fs to 22 fs is demonstrated experimentally in a petawatt laser beam.**

Keywords: *femtosecond laser pulses, temporal compression, self-phase modulation.*

1. Introduction

The present-day knowledge and technologies enable designing sources of laser radiation with a peak power exceeding 1 PW [1, 2]. Focusing of such radiation allows a record peak intensity up to 10^{22} W cm^{-2} to be achieved [3]. Ultra-high-power optical pulses are used to study the behaviour of matter in ultra-intense light fields and to accelerate elementary particles pulled out by the field from gas and solid targets [4]. The next step in the application of high-power lasers may be investigation of nonlinear properties of vacuum and processes of the birth of electron–positron pairs [5]. This demands substantial enhancement of laser radiation peak power. To this end, the energy of the pulse should be increased and its duration should be reduced. The first method is costly and requires the development of technologies of fabricating wide-band laser amplifiers with an aperture over 10 cm and the corresponding sources for optical pumping. At the same time, it is generally accepted that a minimal duration of petawatt power laser pulses is determined by the gain band of laser amplifiers and the accuracy of stretcher–compressor alignment, as well as by the spectral band of mirrors and the size of diffraction gratings.

Note that today experimentalists have an opportunity to work with petawatt power pulses with a duration over 27–30 fs [6, 7] with an unfocused radiation intensity of several TW cm^{-2}. The pulse duration of high-power radiation may be reduced substantially using the method of additional temporal compression [8, 9]. This method is based on the self-phase modulation effect arising during propagation of intense optical pulses in media with cubic nonlinearity and resulting in spectrum broadening. Correction of the spectral phase of such radiation by means of chirped mirrors reduces significantly the duration [9, 10]. As nonlinear media, 1-mm-thick glass or fused silica plates, as well as transparent polymers are used [9, 11]. This approach allows a several-fold increase in the peak power of femtosecond radiation by using passive elements.

In this paper we consider the impact of residual spectral phase on the processes of spectrum broadening of femtosecond pulses and their further temporal compression. We present experimental results on spectrum broadening and subsequent temporal compression of part of a petawatt power beam. The experiments have been performed on the PEARL laser facility [12].

2. Impact of residual phase on spectrum broadening in media with cubic nonlinearity

Let us consider spectrum broadening and subsequent temporal compression of intense (~TW cm^{-2}) femtosecond pulses with spectral phase modulation. Phase modulation may be produced artificially as a result of mismatched operation of a stretcher and a compressor or may be a consequence of technical impracticability of full spectral phase correction [13].

The problem is formulated as follows. At the input of a nonlinear medium there is an intense laser pulse with phase modulation. We will denote the envelope amplitude of the electric field by $A(t)$. In the course of propagation in the medium, the pulse acquires additional nonlinear phase modulation and broadens its spectrum. The modification of pulse parameters is described by the quasi-optical equation in the second approximation of the dispersion theory [14]:

$$\frac{\partial A}{\partial z} + \frac{1}{u}\frac{\partial A}{\partial t} - \mathrm{i}\frac{k_2}{2}\frac{\partial^2 A}{\partial t^2} + \mathrm{i}\gamma |A|^2 A = 0, \quad (1)$$

where $\gamma = 3\pi k_0 \chi^{(3)}/(2n_0^2)$; u is the group velocity; z is the longitudinal coordinate; $k_2 = \partial^2 k/\partial\omega^2|_{\omega_0}$ is the parameter of group velocity dispersion; n_0 is the linear part of the refractive index; k_0 is the wave vector; and $\chi^{(3)}$ is the cubic nonlinear susceptibility. The influence of cubic polarisation is characterised by the accumulated nonlinear phase (B-integral): $B = \gamma |A_{max}|^2 L$, where L is the length of a medium and A_{max} is the maximum value of the field amplitude. Note that the B-integral values smaller than unity do not lead to an appreciable spectrum distortion and will not be considered in the present paper.

External quadratic correctors of spectral phase allow the input pulse duration to be reduced. This procedure is described as follows:

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$$A_c(t) = F^{-1}\left(\exp\left(-\frac{i\alpha_{opt}\Omega^2}{2}\right)F(A_{out}(t, z = L))\right). \quad (2)$$

Here, F and F^{-1} are the direct and inverse Fourier transforms; $A_{out}(t, z = L)$ is the field envelope at the nonlinear medium output; Ω is the offset from the centre frequency; and α_{opt} is the parameter providing the highest peak power P_c of the compressed pulse. In this formulation, the negative (positive) sign of the parameter α_{opt} corresponds to anomalous (normal) group velocity dispersion of the phase correctors.

Let us find how the spectral phase of the initial pulse with $A(t, z = 0)$ affects the spectrum broadening processes and subsequent temporal compression with the use of quadratic phase correctors. To do so, we will specify the initial Gaussian pulse spectrum in the form

$$S(\Omega) = S_0 \exp\left[-2\ln 2\frac{\Omega^2}{\Omega^2_{FWHM}} - i\varphi(\Omega)\right]. \quad (3)$$

Here, S_0 is the spectrum amplitude and Ω_{FWHM} is the FWHM spectrum width. As the quadratic component of spectral phase can always be corrected by an optical compressor, we will restrict our consideration to the residual phase of form

$$\varphi(\Omega) = \frac{\beta}{6}\Omega^3 + \frac{\delta}{24}\Omega^4.$$

In the temporal domain this spectrum distribution corresponds to the pulse with an amplitude

$$A(t) = \int_{-\infty}^{\infty} S(\Omega)\exp(i\Omega t)\,d\Omega. \quad (4)$$

We use for modelling the following pulse parameters: centre wavelength 910 nm, Fourier limit duration T_F = 57 fs (FWHM), and spectrum width Ω_{FWHM} = 22 nm. Let us take as a nonlinear sample a 0.5-mm-thick polyethylene terephthalate plane-parallel plate with $k_2 = 120$ fs^2 mm^{-1}. We will restrict our consideration to the range of the parameters $\beta \in [-7; 7] \times 10^4$ fs^3, $\delta \in [-3; 3] \times 10^6$ fs^4, where either cubic phase modulation or fourth-order modulation leads to a 20 % reduction of peak intensity.

The efficiency of the process of peak power enhancement will be characterised by the parameter $K = P_c/P_{in}$ defined by the ratio of the compressed pulse peak power P_c to the initial pulse peak power P_{in}. Introduction of spectral phase modulation leads to a pulse shape distortion, peak intensity reduction and, as a consequence, to a reduced value of the accumulated B-integral. Therefore, for a more correct comparison of the potential peak power enhancement due to spectrum broadening and further temporal compression, we will assume that with a change in parameters β and δ, the accumulated nonlinear phase remains constant due to an increase in the input pulse energy. The energy of the pulses with the values of β or δ from the considered range should be increased by no more than 1/0.8 = 1.25 times. The values of K obtained as a result of quadratic phase correction of the spectrum using formula (2) are presented in Fig. 1a for the B-integral equal to 3.1. For a fixed value of the B-integral, peak power increase is the largest for pulses with frequency phase modulation at positive values of the parameter δ and values of the parameter β close to zero, rather than for a transform-limited pulse at the nonlinear medium input. An asymmetric dependence of the parameter K on the sign of the parameter δ is typical for media with $\gamma > 0$ and $k_2 > 0$.

The distribution diagram of the optimal value of the quadratic spectral phase correction, α_{opt}, is presented in Fig. 1b. The parameter was optimised by the highest peak power after spectrum broadening and quadratic spectral phase correction. Despite a rather wide range of variation of β and δ, a maximum deviation of α_{opt} does not exceed 18 % of the average value.

The diagram for the coefficient of peak power increase, K_{FL}, at full spectral phase correction of the pulse that has passed through a nonlinear medium, i.e. for the Fourier-transform-limited (FTL) pulse with $A_{out}(t)$, is shown in Fig. 1c. Comparison of Figs 1a and 1c shows that full spectral phase correction provides an insignificant gain in peak power, namely, by a factor of 2.9 against a factor of 2.6. Consequently, correction of the quadratic phase component only is quite sufficient for experimental implementation. Note that full correction of the spectral phase after spectrum broadening is important for enhancement of the temporal intensity profile and helps localise energy at the main peak.

It should be emphasised that, within the framework of the considered approximation, the coefficient of peak power increase, K, at fixed values of parameters β and δ depends almost linearly on the B-integral. It is also worthy of notice that an increase in linear dispersion k_2 of the material does not change the form of the curve (see Fig. 2). An analogous dependence of K on the B-integral was observed when the initial pulse had only a cubic phase [15].

This method of peak power enhancement has some physical limitations. The first limitation is caused by the laser dam-

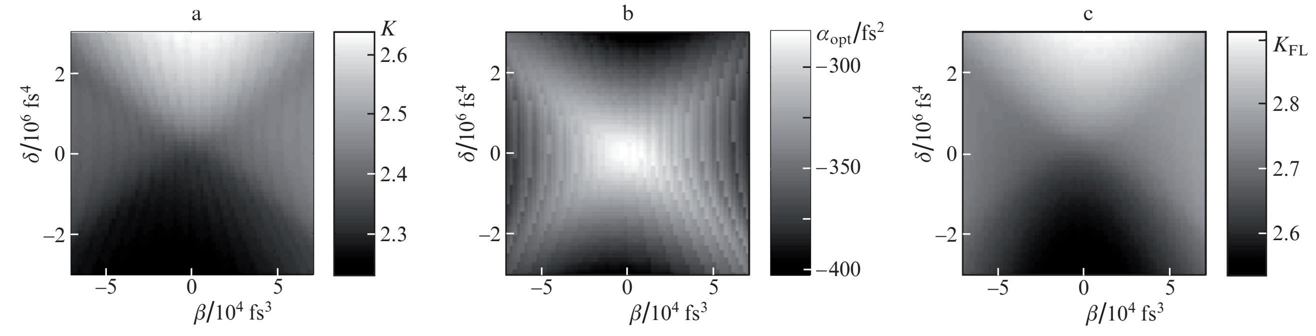

Figure 1. Distribution diagrams of (a) peak power increase coefficient K after quadratic spectral phase correction, (b) optimal parameter α_{opt} and (c) peak power increase coefficient K_{FL} after full spectral phase correction vs. parameters β and δ.

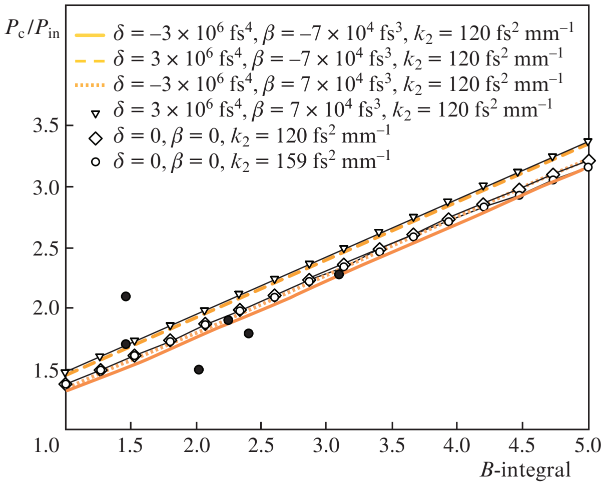

Figure 2. Calculated peak power increase vs. B-integral for different values of parameters β, δ and k_2 (grey lines and empty symbols); filled circles show the experimental data.

age of the transmissive optical element as a result of developing small-scale self-focusing (SSSF) for the B-integral values exceeding unity. The method of SSSF suppression that may be used for studying petawatt power radiation was considered in [16]. The second limitation arises due to the need to take into account inertia of the nonlinear response of cubic polarisation, as a result of which the group velocity becomes intensity-dependent, the pulse tail (front) steepens at $\gamma > 0$ ($\gamma < 0$), and an envelope shock wave is formed if the medium is thick enough [14]. Such effects occur as a rule for intense pulses with 5–10 fs initial durations that will not be considered here.

3. Temporal compression of intense pulses with frequency phase modulation in the experiment

The processes of spectrum broadening and subsequent temporal compression by means of chirped mirrors were studied in experiments on the subpetawatt PEARL laser facility. An industrial polyethylene terephthalate sample 0.5 mm thick was taken as a nonlinear medium where self-phase modulation and, as a result, broadening/modification of the frequency spectrum occurred. This material was chosen for the following reasons: unlimited aperture with respect to laser beams (> 100 mm) for sample thickness less than 1 mm, high enough quality for transmissive optics [11], and low cost.

The schematic of the experiment is presented in Fig. 3. The diaphragm (*1*) cut out a 20 mm region from a 100-mm beam. The diaphragm was used to reduce the beam size so as to use chirped mirrors with a 2 inch aperture for pulse shortening. (Note that the diaphragm is not needed for practical applications; in this case the size of chirped mirrors will depend on the laser beam size and the mirrors should be placed inside a vacuum chamber.) A 0.5-mm-thick plane-parallel polyethylene terephthalate element placed immediately behind the diaphragm partially overlapped the passing beam. For further temporal compression and control of parameters, the optical pulse was attenuated by a factor of 100 as a result of reflection from the front surface of the wedge (*3*). The pulse spectrum was measured by an image-spectrograph. For measurements of duration we used a single-shot intensity autocorrelator. As the plane-parallel plate overlapped part of the laser beam, we measured during a single short the spectrum and autocorrelation function (ACF) of both, the initial pulse and the pulse that had passed through the nonlinear sample and temporal compression system.

In our experiments we used laser pulses with the following parameters: centre wavelength 920 nm, pulse energy 5.5 J and beam size ~100 mm. The intensity distribution in the beam was quasi-plane with a fill factor ~0.7. In this case, spectrum broadening across the laser beam is quite uniform and does not depend on the position of the beam. The measured spectrum and autocorrelation function of the initial pulse intensity are shown by solid curves in Figs 4a and 4b, respectively.

Note that the spectral phase was not measured in our experiments. The problem of spectral phase retrieval from the measured spectrum and ACF has no unambiguous solution. At the same time, the influence of the residual quadratic spectral phase on the duration of high-power pulses (without

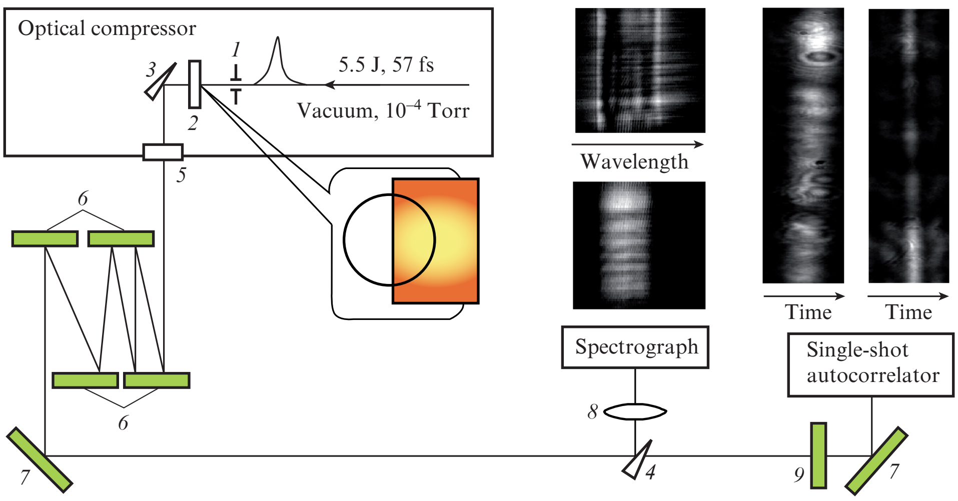

Figure 3. Schematic of the experiment on additional temporal compression: (*1*) 20-mm-diameter diaphragm; (*2*) 0.5-mm-thick plane-parallel polyethylene terephthalate sample; (*3, 4*) 1-mm-thick silica glass wedges; (*5*) 4-mm-thick K8 glass compressor output window; (*6*) chirped mirrors with anomalous dispersion (–100 fs^2); (*7*) 45° dielectric mirror; (*8*) spherical lens; (*9*) dispersion compensator.

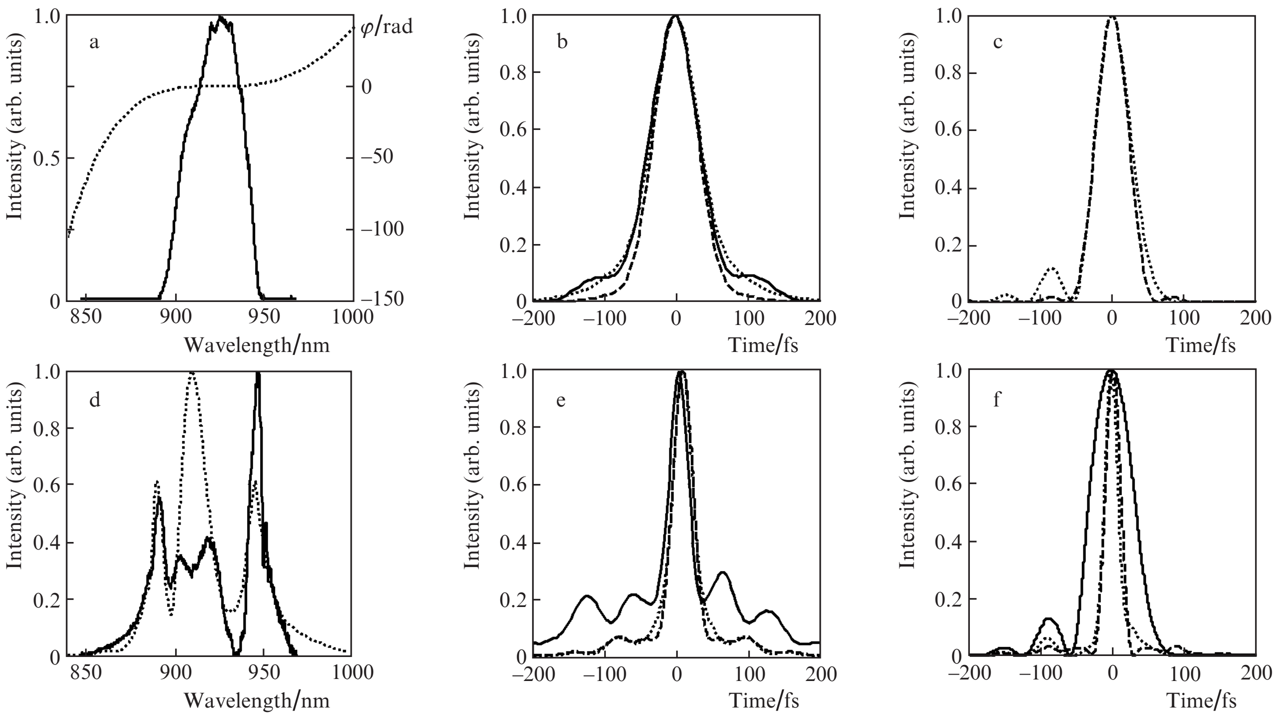

Figure 4. Spectra, ACFs and profiles (a–c) in front of the nonlinear sample and (d–f) behind it: (a) measured spectrum (solid curve) and fitted spectral phase φ (dotted curves); (b) intensity ACF of the initial pulse in the experiment (solid curve), of the FTL pulse calculated from the spectrum (dashed curve) and fitted (dotted curve); (c) profiles of the fitted pulse (dotted curve) and the FTL pulse from the spectrum (dashed curve); (d) experimental (solid curve) and numerically modelled (dotted curve) broadened spectra; (e) intensity ACF of the compressed pulse in the experiment (solid curve), the pulse after spectral phase correction at $\alpha = -238$ fs^2 (dotted curve) and the FTL pulse from the experimentally broadened spectrum (dashed curve); (f) result of numerical modelling of pulse shapes at the nonlinear medium output (solid curve), after correction of the spectrum quadratic phase at $\alpha = -238$ fs^2 (dotted curve) and the FTL pulse from the experimentally broadened spectrum (dashed curve).

propagation through the nonlinear sample) was minimised by aligning the optical compressor. Consequently, we sought for the spectral phase like in the theoretical part of the study: $\varphi(\Omega) = (\beta/6)\Omega^3 + (\delta/24)\Omega^4$. Using numerical methods we found that the parameters $\beta = -6.91 \times 10^4$ fs^3 and $\delta = -2.1 \times 10^4$ fs^4 ensured rms deviation of the calculated ACF from the measured one (see Fig. 4b). The pulse shape corresponding to the spectral phase is presented in Fig. 4c. The pulse duration is 57 fs. The durations of the measured and calculated ACFs are, respectively, 79 fs and 78 fs. At the same time, the duration of the FTL pulse corresponding to the spectrum in Fig. 4a is 53 fs.

During propagation in a material medium (a 4-mm-thick compressor output window, 1 m air, 1 mm wedge, complementary dispersion compensator which is a polyethylene terephthalate plate 1.5 mm thick), the optical pulse accumulates positive frequency group velocity dispersion. For the centre wavelength of 920 nm, the material of the K8 window, fused silica and air have the following linear dispersions: $k_2 = 33.8$, 28.6 and 18×10^{-3} fs^2 mm^{-1}. The dependences of refractive indices on wavelength for fused silica and glasses were borrowed from [17], and for air from [18]. The total positive dispersion accumulated by the pulse in the course of propagation from the nonlinear sample to the single-shot autocorrelator was 362 fs^2. The spectral phase was corrected by means of dielectric chirped mirrors (UltraFast Innovations GmbH, Germany) with anomalous dispersion of -100 fs^2 introduced at each reflection. Six reflections from chirped mirrors were used to achieve temporal compression, so that the total anomalous dispersion was -600 fs^2. Thus, the residual anomalous dispersion $\alpha_e = -238$ fs^2 was used for correcting the spectral phase accumulated in the 0.5-mm-thick polyethylene terephthalate sample.

The ratio of the ACF initial duration (T_{ACF}^{in}) to the ACF after correction of the quadratic spectral phase (T_{ACF}^{c}) versus parameter α [see formula (2)] is plotted in Fig. 5. The curve was calculated for the parameters of the experiment. The experimental point is close to the optimal position in the graph (-284 fs^2). It is also worth noting that the curve in

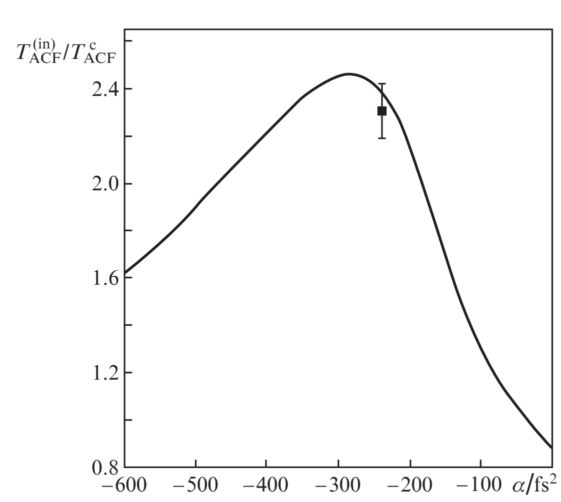

Figure 5. Ratio of the initial ACF to the ACF after spectral phase correction vs. quadratic spectral phase correction parameter α (solid curve); the filled point is the experiment.

Fig. 5 is rather smooth. Hence, even a major error in selecting the spectral phase correction parameter α does not lead to a significant decrease in the compressed pulse power. For example, an error of 100 fs^2 relative to the optimal value results in reduction of the coefficient of peak power increase from 2.4 to 2.1.

The spectra broadened after passage through the nonlinear element obtained in the experiment and in numerical modelling are presented in Fig. 4d. The spectra topologies agree qualitatively: broadening relative to the initial spectrum is observed and narrow peaks are formed. However, the peak scale and amplitude are different in the numerical modelling and experiment. A possible reason is ambiguity in determining the phase of the initial spectrum. The pulse shapes obtained in the numerical modelling at the nonlinear medium output after quadratic phase correction at $\alpha = -238$ fs^2, as well as for the Fourier limit of the spectrum are shown in Fig. 4f. The pulse durations are, respectively, 68, 22 and 21 fs. The ACFs obtained in experiment and in numerical modelling are plotted in Fig. 4e. The durations of the calculated and experimental ACFs are 33 and 34 fs, respectively. Thus, as a result of compression obtained in the experiment using chirped mirrors, the duration of the autocorrelation function (pulse) was reduced from 79 (57) fs to 34 (22) fs.

For the considered parameters, addition of the self-steepening phenomenon to Eqn (1) changes the dependences negligibly in Figs 1 and 4. Note that the compressed pulse has a sufficiently strong pedestal near the main maximum. It is, evidently, the result of incomplete correction of spectral phase before the nonlinear sample and after the compression stage. The pulse profile could be improved by using a feedback between DAZZLER or AOPDF (acousto-optic programmable dispersive filter) and SPIDER (spectral phase interferometry for direct electric-field reconstruction) devices, but it is difficult to obtain the feedback in experiments with a low repetition rate (1 shot per 40 min) lasers like PEARL.

The peak intensity in the medium reached $\sim$1.5 TW cm^{-2} and the accumulated B-integral was $\sim$3.1. For estimating the B-integral we used the value of cubic nonlinearity of 6×10^{-7} cm^2 GW^{-1} [11].

Note that the ACF of the initial pulse was measured in the beam region that was outside the nonlinear medium but experienced six reflections from the chirped mirrors. Notwithstanding this fact, the error in such measurements of the initial pulse duration was insignificant for the considered parameters of the experiment, as addition to the initial spectrum only of quadratic phase with the frequency dispersion parameter of -238 fs^2 (Fig. 4a) increases the duration as compared to the Fourier limit of 53 fs by less than 0.5 fs.

It should be emphasised that due to the use of quasi-flat top laser beams (the fill factor of about 0.7) in the experiments, the transverse distribution of the accumulated B-integral is quasi-uniform. As a result, the spectrum broadens identically and the impact of the acquired wave front distortions is insignificant. Also, the distribution of the pulse duration should be quasi-uniform across the beam. This will be verified in the theoretical and experimental research to follow. A possibility of using the technique for laser pulses with an inhomogeneous (quasi-Gaussian) intensity distribution was described in [19–21].

It is worthy of notice that the used approach to peak power enhancement of optical pulses can be quite readily adapted for use in the front end of ultra-high-power lasers. That was verified by the experimental data (see Fig. 2) on pulse compression at the output of the front end of the PEARL laser facility. In our experiment we used laser radiation with a centre wavelength of 910 nm, a beam diameter of $\sim$20 mm, a fill factor of $\sim$0.5, a pulse energy of $\sim$20 mJ and a duration of 66 fs (a 50-fs FTL pulse). A 3-mm-thick polyethylene terephthalate sample was used for spectrum broadening. The spectral phase was corrected due to reflection from the system of chirped mirrors with a total dispersion of -600 fs^2. For the parameters of the study, the assessed value of the B-integral was $\sim$2.2. The pulse duration in the experiment was reduced down to 31–34 fs.

4. Conclusions

We have presented results of numerical modelling of the influence of residual phase of high-power laser radiation on spectrum modification in thin plane-parallel nonlinear samples and subsequent temporal compression using quadratic spectral phase correctors. It has been demonstrated that, for a fixed value of the B-integral accumulated in the nonlinear sample, maximum peak power enhancement is reached for pulses with $\delta \geqslant 0$, $\beta = 0$. An increase in the accumulated B-integral leads to a linear growth of the peak power. The experiments on additional temporal compression of high-power laser pulses have demonstrated more than a two-fold reduction of duration/peak power enhancement when only passive optical elements are used. The considered technique may be used as a final stage of peak power enhancement in terawatt and petawatt power laser systems, independent of their concepts and design features.

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